NONSTATIONARY CONVECTIVE HEAT AND MASS TRANSFER OF DROPLETS FOR COMMENSURARE PHASE RESISTANCES

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The linear problem of convective mass and heat transfer of droplets for commensurate phase resistances was investigated in [1-8] for large Peclet numbers. It was considered in [1-7] that a thin diffusion boundary layer exists on both sides of the phase separation; the problem was considered in a stationary formulation in [1, 2, 5]. The solutions obtained in [3, 4, 6, 7] emerged from the stationary regime corresponding to a certain inhomogeneous concentration distribution in the boundary layers which determine a mean Sherwood number different from zero and proportional to $\sqrt{\mathrm{Pe}}$. The complete problem of the mass transfer of drops for commensurate phase resistances was modeled in [8] by the transport equation within the drop with a boundary condition on the surface that reflects the nature of the mass transfer in the continuous phase.

It is shown in this paper that the results of [3, 4, 6, 7] are firstly applicable only in the initial fast-flowing stage of the process (to the start of interaction between the internal diffusion boundary layer and the diffusion wake it generates) and are inapplicable to the intermediate and concluding stages of the process, and secondly, the results of the modeling [8] are reflected poorly in the dynamics of the mass and heat transfer process within the droplet in the initial and intermediate stages of the process but better in the concluding stage.

1. FORMULATION OF THE PROBLEM

Let us consider nonstationary convective mass and heat transfer between droplets and a continuous medium in the case of commensurate phase resistances. Let us consider the phase equilibrium condition in the form

$$C_2 = \alpha F(C_1) \quad (F(0) = 0,\ F'_c \geqslant 0,\ \alpha \geqslant 0), \tag{1.1'}$$

satisfied on a droplet surface, where C_1 and C_2 are the concentrations in the continuous and disperse phases, and $\Psi = \alpha F(C_1) C_1^{-1}$ is the distribution coefficient [8]. It is ordinarily considered that the function F depends linearly on the concentration $F(C_1) = C_1$ [1-7]. It was shown in [8, 9] that a power-law dependence $F(C_1) = C_1^n$ should be used in a number of cases, where $n \approx 0.6$, for instance [9]. The most general case of an arbitrary dependence $F = F(C_1)$ in (1.1') will be examined below.

The appropriate boundary value problem has the following form in dimensionless variables:

$$\partial c_i/\partial t + (\mathbf{v}_i \nabla) c_i = (\varkappa_i/\mathrm{Pe}) \Delta c_i,\ i = 1, 2,\ \varkappa_1 = \varkappa,\ \varkappa_2 = 1; \tag{1.1}$$

$$t = 0,\ c_1 = 1,\ c_2 = 0;\ r \to \infty,\ c_1 \to 1; \tag{1.2}$$

$$r = 1,\ c_2 = f(c_1),\ \varkappa \partial c_1/\partial r = \partial c_2/\partial r, \tag{1.3}$$

$$c_1 = 1 + C_*^{-1}(C_1 - C_{10}), \quad c_2 = C_*^{-1}(C_2 - C_1), \quad \mathrm{Pe} = aUD_2^{-1}, \quad t = a^{-1}Ut',$$
$$\varkappa = D_1 D_2^{-1}, \quad f(c_1) \equiv -C_{20}C_*^{-1} + \alpha C_*^{-1} F(C_*(c_1 - 1) + C_{10}),$$

where C_{10} and C_{20} are the initial concentrations outside and inside the droplet, D_i are diffusion coefficients, Pe is the Peclet number, *a* is the droplet radius, U is the characteristic

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stream velocity, C_* is the characteristic concentration which is selected from considerations of convenience (in the linear case $F(C_1) = C_1$ it is convenient to set $C_* = C_{10} - \alpha^{-1}C_{20}$, then the first boundary condition (1.3) takes the form $r = 1$, $c_2 = \alpha c_1$), t' is the dimensional time. When writing the equations and the initial and boundary conditions (1.1)-(1.3), the quantities a and aU^{-1} were selected as the characteristic length and time scales.

For definiteness we consider the fluid velocity distributions v_i outside and inside the droplet to correspond to the Hadamard–Rybchinskii solution

$$\psi^{(1)} = (1/4)(1 - r)[2(\beta + 1)r - \beta - \beta r^{-1}] \sin^2\theta,\ r \geqslant 1, \tag{1.4}$$

$$\psi^{(2)} = (1/4)r^2(1 - r^2)\sin^2\theta,\ 0 \leqslant r \leqslant 1,$$

$$\mathbf{v}_i = \left(v_r^{(i)}, v_\theta^{(i)}\right),\quad v_r^{(i)} = -\frac{1}{r^2\sin\theta}\frac{\partial\psi^{(i)}}{\partial\theta},$$

$$v_\theta^{(i)} = \frac{1}{r\sin\theta}\frac{\partial\psi^{(i)}}{\partial r}.$$

For convenience, the quantity $U = U_\infty(\beta + 1)^{-1}$, where U_∞ is the stream velocity at infinity and β is the ratio of the droplet viscosity to the surrounding fluid viscosity, was taken as the characteristic stream velocity in writing (1.1)-(1.4); the angle θ is measured from the flow direction at infinity (see Fig. 1).

It should be noted that the limit cases of limiting resistance of the outer or inner phases correspond to passages to the limit in (1.1')-(1.4) as $\alpha \to \infty$ and $\alpha \to 0$. Furthermore, the most important case of commensurate phase resistances will be chosen, for example, that which occurs in the corresponding thermal problem where the equalities $\alpha = 1$, $F(C_1) = C_1$ are always satisfied in (1.1') (here C_1 and C_2 are the temperature outside and inside the drop).

An asymptotic analysis of the problem (1.1)-(1.4) for large Peclet numbers (Pe >> 1) (it is considered that $f(c_1) = O(1)$ for $c_1 = O(1)$) shows that the flow core e_i, the diffusion boundary domain layer domain d_i, and the diffusion wake domain W_i, $i = 1, 2$ (see sketch) can be extracted in the whole flow domain outside and inside the drop.

2. INITIAL STAGE OF THE PROCESS

For $t \ll 1$ the convective term in (1.1) can be neglected, and the solution will here correspond to a thin "shock" spherical layer outside and inside the droplet near its surface (which is a result of the mismatch between the initial and boundary conditions (1.2) and (1.3) for $f(1) \neq 0$).

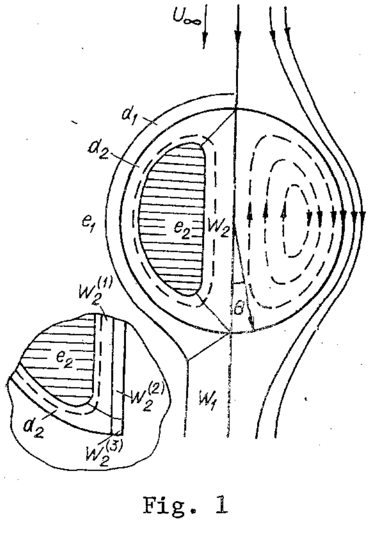

Fig. 1

For $t = O(1)$ the convective terms in (1.1) will be of the same order as the nonstationary terms; hence the concentration in the stream cores remains equal to the initial concentrations $c_1^{(e)} = 1$ and $c_2^{(e)} = 0$, while diffusion boundary layers $d_i = \{|r-1| \leqslant O(\mathrm{Pe}^{-1/2}),\ O(\mathrm{Pe}^{-1/2}) \leqslant \theta,\ O(\mathrm{Pe}^{-1/2}) \leqslant \pi - \theta\}$ are formed near the droplet surface (here and henceforth, the inequalities in the braces indicate the orders of the characteristic dimensions of the domain under consideration), which still behave qualitatively identically on different sides of the droplet surface. The introduction of a new "stretched" boundary layer coordinate Y in the equation, initial and boundary conditions (1.1)-(1.4) with subsequent extraction of the higher terms of the expansion in the small parameter $\mathrm{Pe}^{-1/2}$ results in the following boundary value problem

$$\frac{\partial c_i^{(d)}}{\partial t} = \Lambda^{(i)} \cdot c_i^{(d)}, \quad \Lambda^{(i)} \equiv \varkappa_i \frac{\partial^2}{\partial Y^2} - Y\mu \frac{\partial}{\partial Y} - \frac{1}{2}(1-\mu^2)\frac{\partial}{\partial \mu}, \quad i = 1, 2; \tag{2.1}$$

$$t = 0,\ c_1^{(d)} = 1,\ c_2^{(d)} = 0; \quad Y \to -\infty,\ c_1^{(d)} \to 1; \quad Y \to +\infty,\ c_2^{(d)} \to 0; \tag{2.2}$$

$$Y = 0, \quad c_2^{(d)} = f\left(c_1^{(d)}\right), \quad \varkappa \frac{\partial c_1^{(d)}}{\partial Y} = \frac{\partial c_2^{(d)}}{\partial Y}, \tag{2.3}$$

$$Y = \mathrm{Pe}^{1/2}(1-r), \quad \mu = \cos\theta \quad (\varkappa_1 = \varkappa,\ \varkappa_2 = 1).$$

We seek the solution of (2.1)-(2.3) in the form

$$c_i^{(d)} = c_i^{(d)}(\zeta, \mu, \omega) = A_i + B_i \operatorname{erf}\left\{\frac{|\zeta|}{2\varkappa_i^{1/2}[\sigma(\mu) - \sigma(S(\omega))]^{1/2}}\right\}, \tag{2.4}$$

$$\zeta = \mathrm{Pe}^{1/2}\psi = \frac{1}{2}Y(1-\mu^2), \quad \sigma(\mu) = \frac{1}{6}(2-\mu)(1+\mu)^2,$$

$$\omega = t - \ln\frac{1+\mu}{1-\mu}, \quad S(\omega) = -\operatorname{th}\frac{\omega}{2}.$$

It can be shown that (2.4) is the solution of (2.1) for any constants A_i and B_i, whose explicit form is obtained by substituting (2.4) in the initial and boundary conditions (2.2) and (2.3). We consequently have

$$A_1 = 1 - \lambda\varkappa^{-1/2}, \quad B_1 = \lambda\varkappa^{-1/2}, \quad A_2 = \lambda, \quad B_2 = -\lambda; \tag{2.5}$$

$$\lambda = f(1 - \lambda\varkappa^{-1/2}). \tag{2.6}$$

Here the parameter λ is determined by solving the (transcendental) equation (2.6). In the linear case $f(c) = c$ formulas (2.4)-(2.6) go over into formulas from [4, 6].

It can be shown that the expressions (2.4) become unsuitable in the neighborhood of the outflow point $\theta = 0$ ($\mu = 1$), where the formation of the inner and outer diffusion wakes W_i occurs. In the outer wake W_1 the substance depleted because of diffusion is entrained by the stream from the droplet surface to infinity, and exerts no reverse influence on the outer diffusion boundary layer that generates it. An essentially more complex situation holds within the droplet. For $t = 0$ the concentration distribution within the droplet is homogeneous and the diffusion wake W_2 is missing; for $t > 0$ the diffusion wake starts to be formed and by gradually "progressing" along the stream axis, "penetrates" through the whole volume of the droplet, after which it starts to interact with the internal diffusion boundary layer d_2 by exerting a reverse influence on it.

The diffusion wake W_2 is substantially inhomogeneous and consists of the domain of the rear critical point $W_2^{(3)} = \{1 - r \leqslant O(\mathrm{Pe}^{-1/2}),\ \theta \leqslant O(\mathrm{Pe}^{-1/2})\}$, the convective boundary layer domain of the wake $W_2^{(1)} = \{O(\mathrm{Pe}^{-1/2}) \leqslant 1 - r,\ O(\mathrm{Pe}^{-1}) \leqslant \psi^{(2)} \leqslant O(\mathrm{Pe}^{-1/2})\}$, the internal domain of the wake $W_2^{(2)} = \{O(\mathrm{Pe}^{-1/2}) \leqslant 1 - r,\ 0 \leqslant \psi^{(2)} \leqslant O(\mathrm{Pe}^{-1})\}$, and the domain of the leading stagnation point $b = \{1 - r \leqslant O(\mathrm{Pe}^{-1/2}),\ \pi - \theta \leqslant O(\mathrm{Pe}^{-1/2})\}$ (see Fig. 1). The procedure to obtain the equations and boundary conditions for the concentration distribution in the domains mentioned is completely analogous [10, 11]. The main transfer of substance coming in from the boundary layer occurs in the convective-boundary layer domain of the diffusion wake $W_2^{(1)}$, in which the concentration distribution is defined by the equation

$$\frac{\partial c_2^{(1)}}{\partial t} - \frac{1}{2}(1-z^2)\frac{\partial c_2^{(1)}}{\partial z} - \frac{1}{2} z\eta \frac{\partial c_2^{(1)}}{\partial \eta} = 0, \quad \eta = \mathrm{Pe}^{1/4}\rho, \tag{2.7}$$

where $z = r\cos\theta$, $\rho = r\sin\theta$ is a cylindrical coordinate system related to the center of the droplet, and η is the "stretched" coordinate. Equation (2.7) is obtained by inserting the new variables z and η in (1.1) and subsequently extracting the higher terms of the expansion in the small parameter $\mathrm{Pe}^{-1/2}$ (as always, in this procedure it is considered that $\eta = O(1)$, $\partial/\partial\eta = O(1)$, $\partial^2/\partial\eta^2 = O(1)$).

The general solution of (2.7) has the form

$$c_2^{(1)} = \Phi(\zeta, \omega^{(1)}), \quad \zeta = \mathrm{Pe}^{1/2}\psi^{(2)} = \frac{1}{4}\eta^2(1-z^2), \quad \omega^{(1)} = t + \ln\frac{1+z}{1-z}, \tag{2.8}$$

where Φ is an arbitrary function of both arguments ζ and $\omega^{(1)}$.

The condition for asymptotic merging of the solution in the diffusion boundary layer domain (2.4)-(2.6) and the convective boundary layer domain of the diffusion wake (2.8) is written in the form

$$c_2^{(d)}\big|_{\mu\to 1} = c_2^{(1)}\big|_{z\to 1}$$

and governs the explicit form of the function Φ, and therefore, the concentration distribution in the domain $W_2^{(1)}$ as well

$$c_2^{(1)} = c_2^{(d)}\left(\zeta, 1, \omega^{(1)} - \frac{1}{2}\ln\mathrm{Pe} + \ln\zeta - 4\ln 2\right). \tag{2.9}$$

Execution of the merger procedure in this case is most simple to comprehend if one continues through coordinate s, measured along the streamline in the flow where the origin of measuring the coordinate s is from the middle of the droplet section $\theta = \pi/2$ and s is the distance from the surface $\theta = \pi/2$ along the streamline $\psi^{(2)} = \text{const}$, is introduced in place of the two local coordinates μ (in the domain d_2) and z (in the domain $W_2^{(1)}$). It can be seen that the local coordinate is expressed in the diffusion boundary layer d_2 in terms of the continuous coordinate as follows: $\mu = \sin s$ $(0 \leqslant s \leqslant \pi/2)$ while in the convective boundary layer domain $W_2^{(1)}$ the relationship $z = \pi/2 + 1 - s$ $(\pi/2 \leqslant s \leqslant \pi/2 + 2)$ is valid. Furthermore, let us note that the local variables ω and $\omega^{(1)}$ are characteristics of the shortened equations (2.1) and (2.7) for $\partial/\partial Y = \partial^2/\partial Y^2 = \partial/\partial\eta = 0$. Taking into account the equality $(\mathbf{v}\nabla)c_2 = v_s\partial c_2/\partial s$, it is easy to verify that one continuous cyclic variable

$$\overline{\omega} = t - \int_0^s \frac{ds}{v_s}, \quad v_s = |\mathbf{v}|, \tag{2.10}$$

can be introduced in place of the local variables ω and $\omega^{(1)}$ corresponding to the local coordinates μ and z by using which the solutions in the diffusion boundary layer and the convective-boundary layer domain of the wake within the droplet can be written in the form

$$c_2^{(d)} = c_2^{(d)}(\zeta, \mu, \overline{\omega}), \quad c_2^{(1)} = \Phi(\zeta, \overline{\omega}). \tag{2.11}$$

Merging the expressions (2.11) already offers no difficulties (in contrast to (2.4) and (2.8)) and is performed in an elementary manner. Here the direct calculations of ω in the domain $W_2^{(1)}$ with the subsequent passage from the continuous coordinate s to the local coordinate z results in (2.9). The presence of logarithmic terms in the representation (2.9) is due to the singularity in the integrand (2.10) in the neighborhood of the outflow point where the fluid velocity v_s is close to zero and essentially inhomogeneous in ψ^2.

Equation (2.7) corresponds to purely convective substance transfer without change along the streamlines. The concentration profile in $W_2^{(1)}$ is determined by the concentration distribution at the "exit" from the diffusion boundary layer d_2. It is seen from (2.9) that a substantial retardation with the characteristic time $\ln\mathrm{Pe}/2$ occurs in the convective boundary layer diffusion wake $W_2^{(1)}$ (as always, it is considered that $\zeta = O(1)$ in $W_2^{(1)}$), and this retardation is different at different streamlines $\zeta = \text{const}$. Such dispersion of the retardation time is due to the inhomogeneity of the fluid velocity distribution near the outflow stagnation point $\theta = 0$, where as follows from (2.9), there is practically no substance transport near the stream axis ($\zeta \ll 1$), and the retardation is quite large. As the distance increases from the stream axis, the fluid velocity in the neighborhood of the outflow point increases and the retardation diminishes.

For the diffusion wake to be formed completely (i.e., "penetrate" throughout the whole droplet volume) and interaction to start with the diffusion boundary layer d_2, the continuous coordinates s "advancing" along the fixed streamline near the stream axis and "passing" the neighborhood of the forward stagnation point ($\theta \approx \pi$) should be incident in the domain d_2. Here because of the complete identity of the singularities of the fluid velocity field in the neighborhood of the forward ($\theta \approx \pi$) and rear ($\theta \approx 0$) stagnation points, the retardation time should be doubled.

It follows from the analysis performed that the domain of applicability of the solutions (2.4)-(2.6) is limited to the range $0 \leqslant t < t_*$, where t_* is the characteristic retardation time

$$t_* = O(\ln \mathrm{Pe}). \tag{2.12}$$

The characteristic retardation time t_* has a simple physical meaning, namely: Define t_* by the formula

$$t_* = J(\psi^{(2)}) = \oint \frac{ds}{v_s}, \tag{2.13}$$

corresponding to the time during which a fluid element within the droplet will perform a complete rotation along the closed line $\psi^{(2)} = \text{const}$ passing through the diffusion boundary layer domain d_2 and the convective-boundary layer domain of the diffusion wake $W_2^{(1)}$, i.e., in the limit for

$$\mathrm{Pe} \to \infty, \ \psi^{(2)} = \mathrm{Pe}^{-1/2}\zeta, \ \zeta = O(1). \tag{2.14}$$

As a result of the expansion of $J(\psi^{(2)})$ we obtain (2.12) from (2.13) and (2.14).

The results obtained can be interpreted as follows. Since the fluid flow velocity within the droplet is finite, then at the beginning, for short times $t < t_*$ there is a homogeneous concentration arriving from the fluid depths, in the domain of the forward stagnation point b_2. This occurs until the enriched solution incident from the "end" of the boundary layer d_2 into the diffusion wake W_2 (because of influx of substance from outside through the droplet surface), and traversing the whole path near the stream axis reaches the "beginning" of the diffusion boundary layer. The characteristic time of substance transfer in the droplet diffusion layer determines the domain of applicability of the solutions (2.4)-(2.6), which for $t > t_*$ ceases to describe correctly the concentration distribution in the diffusion wake (because of the change in the inleakage conditions).

Formula (2.12) permits making the following remark relative to the qualitative behavior of the fundamental mass transfer characteristics outside and inside the droplet. For very large Peclet numbers such that $\ln \mathrm{Pe} > O(1)$, we have $t_* > O(1)$ from (2.12). The characteristic relaxation time t_p of the diffusion boundary layer solution (2.4)-(2.6) is $t_p = O(1)$ and is less than the characteristic retardation time $t_p < t_*$. This latter means that for very large Peclet number the diffusion boundary layers on different sides of the droplet surface will succeed in being built up earlier than the interaction starts between the internal boundary layer and the inner diffusion wake it generated (i.e., the stationary solution $c_{i\infty}^{(d)} = \lim\limits_{t\to\infty} c_i^{(d)}$ $(\zeta, \mu, \omega(t, \mu))$ can be considered as an intermediate asymptotic of the solution of the general nonstationary problem in the domain d_i). This circumstance adduces a certain meaning to the stationary solutions [1, 2, 5]. It follows from the above that the time dependence of the mean diffusion flow on the droplet surface should have a characteristic flat section corresponding to the intermediate "stationary" regime.

In the intermediate stage of the mass and heat transfer process for $O(\ln \mathrm{Pe}) \leqslant t \leq O(\mathrm{Pe})$ the diffusion boundary layer equations (2.1) still remain valid; however, the solution here is already not self-similar, in contrast to (2.4)-(2.6), since the mutual influence of the diffusion wake W_2 and the boundary layer d_2 within the drop must be taken into account. As a result of the interaction, the internal diffusion wake strongly "blurs" the diffusion boundary layer d_2 (here the boundary layers outside and inside the droplets already start to differ substantially). This gradually results in a significant increase in the thickness of the inner boundary layer with time so that the linear approximation of the stream function near the droplet surface, and therefore (2.1) also, becomes unsuitable for a correct description of the process.

Furthermore, the intermediate time interval is not considered but the asymptotic of the solution of the problem (1.1)-(1.3) will be investigated in the concluding stage of process development for $t \geqslant O(\mathrm{Pe})$.

3. CORRESPONDENCE BETWEEN ASYMPTOTIC EXPANSIONS OF THE SPECIMEN AND THE ORIGINAL

Many qualitative and quantitative features of the asymptotic behavior of nonstationary problems with a small or large parameter can be clarified by application of a Laplace–Carlson transformation because of the following useful remark.

Let there be an (arbitrary) function $x = x(t, \varepsilon)$, dependent on the time and a small (large) parameter ε (to simplify the writing the dependence of the function x on the space coordinates is omitted), which in the limit

$$\varepsilon \to 0, \quad \tau = \delta(\varepsilon)t = O(1) \tag{3.1}$$

is expanded in an asymptotic series in the parameter ε (τ is fixed)

$$\varepsilon \to 0, \quad x \doteq \sum_{n=0}^{\infty} \alpha_n(\varepsilon)\, x_n(\tau), \quad \lim_{\varepsilon \to 0} \frac{\alpha_{n+1}}{\alpha_n} = 0. \tag{3.2}$$

The variable τ corresponds to a "fast" or "slow" time depending on the choice of the kind of function $\delta(\varepsilon)$ (the dependence $\delta(\varepsilon) = \varepsilon^k$ is encountered most often). Expansions of the type (3.2) can naturally be constructed as desired by selecting $\delta(\varepsilon)$ differently. In specific cases when the function $x = x(t, \varepsilon)$ is a solution of a certain boundary value problem, the selection of the dependence $\delta = \delta(\varepsilon)$ raises no difficulties as a rule.

Let us set in correspondence with the function $x(t, \varepsilon)$ the transition $\bar{x}(p, \varepsilon)$ obtained by a Laplace–Carlson transformation

$$\bar{x} = \bar{x}(p, \varepsilon) = \mathrm{L} \cdot x, \quad \mathrm{L} \cdot x = p \int_0^{\infty} e^{-pt} x(t, \varepsilon)\, dt. \tag{3.3}$$

In the transform space, the series

$$\varepsilon \to 0, \quad \bar{x} \doteq \sum_{n=0}^{\infty} \alpha_n(\varepsilon)\, \bar{x}_n(q), \quad q = \frac{p}{\delta(\varepsilon)} \tag{3.4}$$

corresponds formally to the series (3.2) because of the properties of the operator L.

Let us note that the asymptotic expansion of the function $\bar{x} = \bar{x}(p, \varepsilon)$ (3.3) as $\varepsilon \to 0$ and $p = \delta(\varepsilon)q$, $|q| = O(1)$ for a given asymptotic sequence $\alpha_n(\varepsilon)$ is unique [13], and therefore agrees with (3.4). Taking account of the remark made and the relationship (3.1), it follows from the comparison of the series (3.2) and (3.4) that the asymptotic expansion of the function $\bar{x}$ in a series in the transform space as $\varepsilon \to 0$ and $|p| = O(\delta)$, $\delta = \delta(\varepsilon)$ corresponds to the expansion of the original x as $\varepsilon \to 0$ and $t = O(1/\delta)$, and conversely. The mentioned properties of the asymptotic expansions permit elementary transferral of the results obtained in the transform space over to the original (certain general assertions relative to the correspondence of the convergence of the series (3.2) and (3.4) can be obtained by a simple reformulation of the theorem presented in [14]). In application to convective heat and mass transfer problems the above means that "compression" ("extension") of the complex parameter p in the transform space corresponds to "extension" ("compression") of the time t in the space of the originals; here the correspondence $t = O(\mathrm{Pe}^{k}) \leftrightarrow |p| = O(\mathrm{Pe}^{-k})$ is satisfied (the simplest case of a power-law dependence of δ on Pe is presented here).

4. CONCLUDING STAGE OF THE PROCESS

The problem (1.1)-(1.4) takes the following form after a Laplace-Carlson transformation with respect to time

$$
\begin{gathered}
p(u_1 - 1) + (\mathbf{v}_1\nabla)u_1 = \varkappa \mathrm{Pe}^{-1}\Delta u_1 \, (1 < r); \qquad (4.1)\\
pu_2 + (\mathbf{v}_2\nabla)u_2 = \mathrm{Pe}^{-1}\Delta u_2 \; (0 \leqslant r < 1); \qquad (4.2)\\
r \to \infty, \; u_1 \to 1; \; r = 1, \; u_2 = \mathbf{L}\cdot f(c_1), \; \varkappa \partial u_1/\partial r = \partial u_2/\partial r, \qquad (4.3)\\
u_i = u_i(\mathbf{r}, \, p) = \mathbf{L}\cdot c_i(\mathbf{r}, \, t).
\end{gathered}
$$

To analyze the problem (1.1)-(1.3) we use the remark made in Sec. 3 that sets up a correspondence between the asymptotic expansions of boundary value problems for the transforms (4.1)-(4.3) and the originals (1.1)-(1.3). Let us examine the limit case

$$
\mathrm{Pe} \to \infty, \; p = q\mathrm{Pe}^{-1}, \; |q| = O(1), \qquad (4.4)
$$

which will correspond to the behavior of the original c_i for large values of $t = O(\mathrm{Pe})$ because of the results in Sec. 3.

We seek the solutions at the stream cores in the form of a regular expansion in reciprocal powers of the Peclet number ($i = 1, 2, j = 0, 1$)

$$
u_i = u_{i0}^{(e)} + \mathrm{Pe}^{-1} u_{i1}^{(e)} + \ldots, \quad u_{i0}^{(e)}/u_{i1}^{(e)} = O(1), \quad u_{ij}^{(e)} = u_{ij}^{(e)}(r, \theta). \qquad (4.5)
$$

Taking account of (4.4) when we substitute (4.5) into the equation and the boundary conditions (4.1)-(4.3), and extracting terms with identical powers of the Peclet number, we obtain for the first terms of the expansion at the stream cores

$$
\begin{aligned}
&(\mathbf{v}_1\nabla)\, u_{10}^{(e)} = 0, \quad r \to \infty, \quad u_{10}^{(e)} \to 1; && (4.6)\\
&(\mathbf{v}_1\nabla)\, u_{11}^{(e)} = \varkappa\Delta u_{10}^{(e)} - q\left(u_{10}^{(e)} - 1\right), \quad r \to \infty, \quad u_{11}^{(e)} \to 0; && (4.7)\\
&(\mathbf{v}_2\nabla)\, u_{20}^{(e)} = 0; && (4.8)\\
&(\mathbf{v}_2\nabla)\, u_{21}^{(e)} = \Delta u_{20}^{(e)} - q u_{20}^{(e)}. && (4.9)
\end{aligned}
$$

In the outer and inner diffusion boundary layers d_i, a "stretched" variable Y is introduced as before, and the solution is sought in the form of the expansion

$$
\begin{gathered}
u_i = u_{i0}^{(d)} + \mathrm{Pe}^{-1/2} u_{i1}^{(d)} + \ldots, \quad u_{i0}^{(d)}/u_{i1}^{(d)} = O(1), \quad u_{ij}^{(d)} = u_{ij}^{(d)}(Y, \mu), \qquad (4.10)\\
Y = \mathrm{Pe}^{1/2}(1 - r), \; \mu = \cos\theta \; (i = 1, \, 2, \; j = 0, 1).
\end{gathered}
$$

Substituting (4.10) into (4.1)-(4.3) with (4.4) taken into account and then expanding in a series in the small parameter $\mathrm{Pe}^{-1/2}$ results in the boundary layer equations and boundary conditions (the differential operator $\Lambda^{(i)}$ is defined in (2.1)):

$$
\begin{aligned}
&\mathbf{\Lambda}^{(i)}\cdot u_{i0}^{(d)} = 0, \quad i = 1, 2; && (4.11)\\
&\mathbf{\Lambda}^{(1)}\cdot u_{11}^{(d)} = \left(1 - \frac{3}{2}\beta\right)\mu Y^2 \frac{\partial u_{10}^{(d)}}{\partial Y} - \frac{3}{2}\beta Y\left(1 - \mu^2\right)\frac{\partial u_{10}^{(d)}}{\partial \mu} - 2\varkappa\frac{\partial u_{10}^{(d)}}{\partial Y}; && (4.12)\\
&\mathbf{\Lambda}^{(2)}\cdot u_{21}^{(d)} = -\frac{1}{2}\mu Y^2 \frac{\partial u_{20}^{(d)}}{\partial Y} - \frac{3}{2} Y\left(1 - \mu^2\right)\frac{\partial u_{20}^{(d)}}{\partial \mu} - 2\frac{\partial u_{20}^{(d)}}{\partial Y}; && (4.13)\\
&Y = 0, \quad u_{20}^{(d)} = \mathbf{L}\cdot f\left(c_{10}^{(d)}\right), \quad u_{21}^{(d)} = \mathbf{L}\cdot\left(\gamma c_{11}^{(d)}\right), \quad \gamma = \left.\frac{\partial f}{\partial c_1}\right|_{c_1 = c_{10}^{(d)}}, && (4.14)\\
&Y = 0, \quad \varkappa\frac{\partial u_{1j}^{(d)}}{\partial Y} = \frac{\partial u_{2j}^{(d)}}{\partial Y}, \quad j = 0, 1.
\end{aligned}
$$

In the convective boundary layer domain of the diffusion wake $W_2^{(1)}$ within the droplet, the solution is sought in the form of the expansion

$$u_2 = u_{20}^{(1)} + \mathrm{Pe}^{-1/2} u_{21}^{(1)} + \ldots, \quad u_{20}^{(1)}/u_{21}^{(1)} = O(1), \tag{4.15}$$
$$u_{2j}^{(1)} = u_{2j}^{(1)}(z, \eta), \quad \eta = \mathrm{Pe}^{1/4} \rho,$$

whose terms satisfy the following equations

$$(1 - z^2)\frac{\partial u_{20}^{(1)}}{\partial z} + \eta z \frac{\partial u_{20}^{(1)}}{\partial \eta} = 0, \tag{4.16}$$
$$(1 - z^2)\frac{\partial u_{21}^{(1)}}{\partial z} + \eta z \frac{\partial u_{21}^{(1)}}{\partial \eta} = 2\eta^2 \frac{\partial u_{20}^{(1)}}{\partial z} - 2\frac{\partial^2 u_{20}^{(1)}}{\partial \eta^2}.$$

The equations and boundary conditions (4.6)-(4.9), (4.11)-(4.14), (4.16) should be supplemented by conditions for the asymptotic merging of the solutions on common boundaries [12, 13]: The solutions in the cores of the outer and inner stream e_i should be merged with the solutions of the outer and inner diffusion boundary layer d_i; the solution in the convective boundary layer domain of the inner diffusion wake $W_2^{(1)}$ should be merged with the solution of the boundary layer d_2 and with the solution in the stream core within the droplet e_2. For the main terms in the asymptotic expansions (4.5), (4.10), (4.15), the mentioned merger conditions are written in the form

$$u_{10}^{(e)}(r \to 1) = u_{10}^{(d)}(Y \to -\infty), \quad u_{20}^{(e)}(r \to 1) = u_{20}^{(d)}(Y \to +\infty), \tag{4.17}$$
$$u_{20}^{(e)}(\rho \to 0) = u_{20}^{(1)}(\eta \to \infty), \quad u_{20}^{(d)}(\mu \to \pm 1) = u_{20}^{(1)}(z \to \pm 1).$$

It follows from (4.6), (4.8) that the zeroth terms of the expansion in the outer and inner flow cores depend only on the stream function.

$$u_{i0}^{(e)} = u_{i0}^{(e)}(\psi^{(i)}), \quad i = 1, 2. \tag{4.18}$$

The asymptotic solution of the problem (4.1)-(4.3) will be constructed in sequence by starting from the external stream core $e_1 \to d_1 \to d_2 \to e_2 \rightleftarrows W_2^{(1)}$.

Taking account of (4.18), it follows from the boundary conditions at infinity (4.6), (4.7), and (4.17) that the first terms of the expansion in the stream core outside the droplet have the form

$$u_{10}^{(e)} = 1, \quad u_{11}^{(e)} = 0. \tag{4.19}$$

It is seen by direct substitution that the highest term of the expansion of the outer diffusion boundary layer, which is a solution of (4.11) for i = 1 and satisfies the condition for merger with the solution in the stream core (4.17), (4.19), has the form

$$u_{10}^{(d)} = 1. \tag{4.20}$$

Taking account of the equality $c_{10}^{(d)} = 1$ (which is a result of (4.20)), it can be seen in an analogous manner that the main term of the inner diffusion boundary layer expansion, which is a solution of (4.11) for i = 2 and satisfies the boundary conditions on the droplet surface (4.14), is determined by the expression

$$u_{20}^{(d)} = f(1). \tag{4.21}$$

The concentration distribution in the convective boundary layer domain of the diffusion wake within the droplet $W_2^{(1)}$ is determined by the solution of (4.16) with the boundary condition $|z| \to 1$, $u_{20}^{(1)} = f(1)$ (which is a result of the mendition for the merger of the solutions $u_{20}^{(d)}$ and $u_{20}^{(1)}$ (4.17)) and has the form

$$u_{20}^{(1)} = f(1). \tag{4.22}$$

Because of (4.21) and (4.22), the condition for merging the solutions in the domains d_2, $W_2^{(1)}$, and e_2 (4.17) yields the boundary conditions for the equation in the stream core within the droplet explicitly

$$\psi^{(2)} = 0, \quad u_{20}^{(e)} = f(1). \tag{4.23}$$

Equation (4.8), whose general solution has the form (4.18), and the boundary condition (4.23) turn out to be insufficient for the determination of the concentration field in the stream core. To obtain the necessary additional information about the function $u_{20}^{(e)}$ we use the equation for the next term in the expansion (4.9). For convenience in the analysis, we introduce a new orthogonal coordinate system ξ, η, φ, connected to the streamlines (the fixed curves η = const are orthogonal to the streamlines $\psi^{(2)}$ = const), which was used in [15]:

$$\xi = 16\psi^{(2)} \equiv 4r^2(1-r^2)\sin^2\theta, \quad \eta = \frac{r^4\cos^4\theta}{2r^2-1},$$
$$g_{\xi\xi} = \frac{1}{64r^2\sin^2\theta\Xi}, \quad g_{\eta\eta} = \frac{(2r^2-1)^4}{16r^6\cos^6\theta\Xi}, \quad g_{\varphi\varphi} = r^2\sin^2\theta,$$
$$\Xi = \Xi(r, \theta) = (1-r^2)^2\cos^2\theta + (2r^2-1)^2\sin^2\theta, \; g = g_{\xi\xi}\, g_{\eta\eta}\, g_{\varphi\varphi}$$
$$(0 \leqslant \xi \leqslant 1, \; -\infty < \eta < +\infty).$$

Equation (4.9) takes the following form in the coordinate system ξ, η, φ,

$$\frac{\partial u_{21}^{(e)}}{\partial \eta} = \left[\frac{\partial}{\partial \xi}\left(\frac{\sqrt{g}}{g_{\xi\xi}}\frac{\partial u_{20}^{(e)}}{\partial \xi}\right) + \frac{\partial}{\partial \eta}\left(\frac{\sqrt{g}}{g_{\eta\eta}}\frac{\partial u_{20}^{(e)}}{\partial \eta}\right)\right] - q\sqrt{g}\,u_{20}^{(e)}. \tag{4.24}$$

The procedure used henceforth is analogous to the sequential elimination of secular (growing) terms when applying the method of stretched coordinates and the method of many scales [12, 13]. Because of the continuity of the solution of the initial problem (1.1)-(1.3) along the fixed streamline ξ = const, the equality

$$c_2(\xi, \eta = -\infty) = c_2(\xi, \eta = +\infty), \tag{4.25}$$

should be satisfied, which means that different points $(\xi, -\infty)$ and $(\xi, +\infty)$ in the new ξ, η coordinate system correspond to the same point $(1/\sqrt{2}, \theta)$ in the old spherical r, θ coordinate system. Integration of (4.24) over φ $(-\infty < \varphi < +\infty)$ with (4.25) taken into account (analogous relationships for all terms of the expansion of the transform of the solution at the stream cores are a result) will result in the following ordinary differential equation

$$\frac{d}{d\xi}\left[\Gamma(\xi)\frac{du_{20}^{(e)}}{d\xi}\right] - qJ(\xi)u_{20}^{(e)} = 0, \quad \xi = 0, \quad u_{20}^{(e)} = f(1), \tag{4.26}$$
$$\Gamma(\xi) = \frac{2}{3}\sqrt{1+\sqrt{\xi}}\left[(4-3\xi)E\left(\sqrt{\frac{1-\sqrt{\xi}}{1+\sqrt{\xi}}}\right) - (4\sqrt{\xi}-3\xi)\times\right.$$
$$\left.\times K\left(\sqrt{\frac{1-\sqrt{\xi}}{1+\sqrt{\xi}}}\right)\right], \quad J(\xi) = \frac{1}{8}\frac{1}{\sqrt{1+\sqrt{\xi}}}K\left(\sqrt{\frac{1-\sqrt{\xi}}{1+\sqrt{\xi}}}\right).$$

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Here E and K are the complete elliptic integrals of the first and second kind, respectively.

The coefficients of (4.26) have the following singularities $\xi \to 0$, $J(\xi) = O(\ln \xi)$; $\xi \to 1$, $\Gamma(\xi) = O(1-\xi)$. A local analysis of (4.26) shows that the first derivative of $u_{20}^{(e)}$ is finite at zero, while the second is infinite and has a logarithmic singularity; the second singularity at the point of degeneration $\xi = 1$ determines two linearly independent solutions, one of which is not bounded as $\xi \to 1$. This latter means that a condition of boundedness of the solution at $\xi = 1$ should still be imposed for equation (2.26) in addition to the boundary condition (4.23).

Expansion of the solution of problem (4.26) as $\psi^{(2)} \to 0$ has the form $u_{20}^{(e)} = f(1) + A\psi^{(2)} + o(\psi^{(2)})$ (where $A = A(q)$ is the transform of the mean concentration at the stream core). Hence, taking account of (4.20) and the second equality of (4.19), the boundary conditions on the droplet surface (4.14) ($j = 1$) and at infinity (corresponding to the asymptotic expansion in the domains e_1, d_1 and e_2, d_2) have the following form for the second terms of the expansions in the inner and outer diffusion boundary layer domains

$$Y=0,\quad u_{21}^{(d)}=\gamma u_{11}^{(d)},\quad \varkappa\frac{\partial u_{11}^{(d)}}{\partial Y}=\frac{\partial u_{21}^{(d)}}{\partial Y}\quad \left(\gamma=\frac{\partial f}{\partial c_1}\Big|_{c_1=1}\right),$$
$$Y\to-\infty,\ u_{11}^{(d)}\to 0;\quad Y\to+\infty,\ u_{21}^{(d)}\to\frac{1}{2}AY(1-\mu^2). \tag{4.27}$$

Equations (4.12) and (4.13), whose right sides vanish because of (4.20) and (4.21), determine, with the boundary conditions (4.27), the second terms of the expansions in the outer and inner boundary layers of the droplets.

It should be noted that the highest term of the expansion in the stream core which is determined by solving the problem (4.26) is uniformly suitable (in high Peclet numbers) in the whole flow domain within the droplet because of (4.21) and (4.22). Consequently, in place of $c_{20}^{(e)}$ we shall simply write c_2.

The reverse transition from the transform to the original results in the following nonstationary equation for the concentration distribution within the drop (t' is the dimensionless time)

$$J(\xi)\frac{\partial c_2}{\partial \tau}=\frac{\partial}{\partial \xi}\Gamma(\xi)\frac{\partial c_2}{\partial \xi},\quad \tau=\mathrm{Pe}^{-1}t\equiv a^{-2}D_2t',$$
$$\tau=0,\ c_2=0;\ \xi=0,\ c_2=f(1);\ \xi=1,\ c_2<\infty. \tag{4.28}$$

For f(1) = 1 Eq. (4.28) agrees exactly with the equation for the purely internal problem examined in [15]. Hence, we have for the mean concentration corresponding to the solution (4.28)

$$\langle c_2\rangle=f(1)\left[1-\frac{3}{2}\sum_{k=1}^{\infty}\alpha_k\exp(-\lambda_k\tau)\right], \tag{4.29}$$
$$\alpha_1=0.4554,\ \alpha_2=0.0654,\ \alpha_3=0.0542,\ \alpha_4=0.0412,\ \alpha_5=0.0038,$$
$$\lambda_1=26.844,\ \lambda_2=137.91,\ \lambda_3=315.66,\ \lambda_4=724.98,\ \lambda_5=1205.2.$$

Here the coefficients α_n and λ_n are presented from data in [8]; for n = 1, 2 values of these coefficients close to those written down were calculated in [15].

5. DISCUSSION OF THE RESULTS. SOME GENERALIZATIONS

Let us note the following circumstances:

1) The solution (2.4)-(2.6) which was obtained in [3, 4, 6] in the linear case $f(c_1)=\alpha c_1$ is valid only in the initial time interval $0\leqslant t<O(\ln \mathrm{Pe})$ (which also determines the domain of applicability of the results [7]);

2) Equation (4.28) corresponding to the nonstationary mass transfer of the drop and the continuous medium for commensurate phase resistances (1.1)-(1.3) is valid only in the concluding stage of process development for $t\geqslant O(\mathrm{Pe})$ and agrees in form (for f(1) = 1) with the Kronig and Brink equation [15] obtained for the purely internal problem;

3) Equation (4.28) is uniformly suitable (in high Peclet number) in the whole domain within the droplet up to its boundary (including the diffusion wake domain also).

The validity of (4.28) for $t\geqslant O(\mathrm{Pe})$ means physically that for sufficiently large times ($t\geqslant O(\mathrm{Pe})$) a stationary regime is actually set up outside the droplet, that corresponds to a homogeneous concentration distribution in the stream while a substantially nonstationary process proceeds inside the droplet when a constant concentration is maintained on its surface, determined by the phase equilibrium condition (1.1') with a stationary homogeneous concentration field outside the droplet. It moreover follows from (4.22) that the concentration on the stream axis within the droplet is already equal, for $t\geqslant O(\mathrm{Pe})$ to the concentration on its surface (because of the multiple circulation of the substance dissolved in the fluid over the closed streamlines).

It should be noted that although the equation and boundary conditions (1.1)-(1.3) were formulated for the concentration problem, all the results obtained are easily carried over to the appropriate thermal problem $c_1 \equiv T_1$, $c_2 \equiv T_2$ (T_1 and T_2 are the dimensionless temperature outside and inside the droplet), which differs from (1.1)-(1.3) only by the boundary conditions (1.3), which have the following form in this case

$$r = 1, \quad T_1 = T_2, \quad (\lambda_1/\lambda_2)\,\partial T_1/\partial r = \partial T_2/\partial r \quad (T_* = T_{10} - T_{20}),$$

where λ_1 and λ_2 are heat conduction coefficients of the fluid outside and inside the droplet. In particular, (4.29) remains valid for $\langle c_2 \rangle \equiv \langle T_2 \rangle$ and $f(1) = 1$ for the mean temperature distribution within the drop for $t \geqslant O(\mathrm{Pe})$.

Let us now investigate the behavior of the solution of problem (1.1)-(1.3) as a function of changes in the ratio between the viscosities of the droplet and the surrounding fluid β. An analysis made corresponded to the case $\beta = O(1)$, and consequently, the final results were independent of the parameter β. In the limit case $\beta \gg 1$ there is practically no flow within the droplet ($\beta = \infty$ corresponds to a solid particle). It can be shown that for $\mathrm{Pe}_\infty = a U_\infty D_1^{-1} \gg 1$ (let us note that Pe_∞ differs by the factor $\varkappa(\beta + 1)$ from the Pe used in (1.1)-(1.3)) and $\tau = a^{-2} D_2 t' \geqslant O(1)$ the concentration (temperature) distribution in the stream outside the particle is homogeneous (in the principal direction, naturally): $c_1 = 1$ and the concentration distribution within the particle is determined by the usual nonstationary heat conduction equation (which corresponds to $v_2 = 0$ in (1.1) for $i = 2$):

$$\frac{\partial c_2}{\partial \tau} = \Delta c_2, \quad \tau = 0, \quad c_2 = 0, \quad r = 1, \quad c_2 = f(1). \tag{5.1}$$

The solution of the problem (5.1) results in the following expression for the mean concentration inside the droplet [8]:

$$\langle c_2 \rangle = f(1)\left[1 - \frac{6}{\pi^2}\sum_{k=0}^{\infty}\frac{1}{k^2}\exp(-\pi^2 k \tau)\right]. \tag{5.2}$$

Comparing (4.29) and (5.2) shows that the order of the characteristic times of nonstationary mass and heat transfer process build-up is identical for droplets of moderate ($\beta = O(1)$) and ultimately large ($\beta \gg 1$) viscosity and is determined by the quantity $\sim a^2 D_2^{-1}$. However, the damping decrements of the solution as $\tau \to \infty$ (substantially) differ by 2.72 times. This latter means that the presence of fluid circulation within the droplets for $\beta = O(1)$ intensifies the convective mass and heat transfer process as compared to a "solid" drop ($\beta = \infty$), where there is no substance transfer because of circulation. It should also be noted that even a significant increase in the external stream velocity U_∞ ($\mathrm{Pe} \to \infty$) influences the degree of substance extraction from the droplet weakly, which has an upper bound as $\mathrm{Pe} \to \infty$ and is proportional to the damping decrement of the mean concentration (4.29). Hence, the relative increment in the degree of extraction because of an increase in the Peclet number tends to zero as $\mathrm{Pe} \to \infty$.

It should be noted that the results of the asymptotic analysis performed are easily extended to the general case of convective mass and heat transfer of droplets of any shape around which there is an arbitrary two-dimensional incompressible (ideal, viscous, etc.) fluid for commensurate phase resistances. In particular, for $\tau = \mathrm{Pe}^{-1} t = a^{-2} D_2 t' \geqslant O(1)$ the concentration distribution outside the droplet is practically homogeneous and is determined by an appropriate value at infinity, while a nonstationary process, described by the following boundary value problem (the condition of boundedness of the solution inside the droplet is omitted), proceeds inside the droplet

$$J(\psi)\frac{\partial c_2}{\partial \tau} = \frac{\partial}{\partial \psi}\Gamma(\psi)\frac{\partial c_2}{\partial \psi}; \quad \tau = 0, \quad c_2 = 0; \quad \psi = 0, \quad c_2 = f(1),$$
$$\Gamma(\psi) = \oint \frac{\sqrt{g}}{g_{\psi\psi}}\,d\chi, \quad J(\psi) = \oint \sqrt{g}\,d\chi, \quad g = g_{\psi\psi} g_{\chi\chi} g_{\varphi\varphi}. \tag{5.3}$$

Here ψ, χ, φ are an orthogonal system connected with the stream function $\psi = \psi^{(2)}$ governing the flow inside the droplet; the curvilinear coordinate ψ orthogonal to χ is found by solving the equation $(\nabla\psi \cdot \nabla\chi) = 0$ (in the plane case $g_{\varphi\varphi} = 1$ should be used, and in the axisymmetric case $g_{\varphi\varphi} = r^2 \sin^2 \theta$). In those cases when there are several independent domains Ω_j (j = 1, ..., n) with perfectly closed streamlines within the droplet which are bounded by singular (critical) streamlines or surfaces (which are closed on the droplet surface), equation (5.3) should be written down and solved independently in all the domains Ω_j by taking into account that for $\tau \geqslant O(1)$ the very same concentration value of f(1), (an example of such a flow is presented in [7] for j = 4) is "taken off" the drop surface onto the singular streamlines.

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